A STUDY ON THERMAL CONDUCTVITY OF EPOXY/Al_2O_3 COMPOSITES

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CERTIFICATE

*This to certify that the thesis entitled "**A Study on Thermal Conductivity of Epoxy/Al_2O_3 Composites**" being submitted by **Asutosh Panda** for the award of the degree of Master of Technology (Thermal Engineering) of NIT Rourkela, is a record of bonafide research work carried out by him under our supervision and guidance. Mr. Asutosh Panda has worked for more than one year on the above problem at the Department of Mechanical Engineering, National Institute of Technology, Rourkela and this has reached the standard fulfilling the requirements and the regulation relating to the degree. The contents of this thesis, in full or part, have not been submitted to any other university or institution for the award of any degree or diploma.*

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CONTENTS

ABSTRACT

The aim of this study is to heighten and examine thermal properties of epoxy based composites using alumina (Al_2O_3) micro-fillers. This work deals with preparation of epoxy composites with high content of micro-filler (Al_2O_3) up to 22.1vol. %.Experimental thermal conductivity tests were evaluated in each specimen and compared with the proposed model. This study was mainly focused on alumina particles of spherical shape is an effective way to enhance both thermal conductivity and sufficient voltage endurance. A numerical simulation using finite element package ANSYS is used to explain heat transfer process within epoxy matrix filled with micro alumina and its effective thermal conductivity values is validated with experimental results and theoretical model correlations. It was elucidated that with 11.3 vol% micro-alumina filled epoxy composites in numerical analysis its thermal conductivity is 0.7$Wm^{-1}k^{-1}$while in experimentation with 22.1 vol% its thermal conductivity is 0.82 $Wm^{-1}k^{-1}$ which is reasonably higher compared with neat epoxy resin. The results show that theAl_2O_3 particles show a percolation behavior at this volume fraction (16vol %) at which a sudden jump in the thermal conductivity is noticed. This is the critical concentration at which Al_2O_3 particles start contacting with each other and hence the actual size of the agglomerates becomes larger. *The effect of the filler size, filler loading, and dispersion conditions of the micro-fillers on the glass-transition temperature (Tg) have been studied.*

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CHAPTER 1

INTRODUCTION

In composites, materials are combined in such a way as to enable us to make better use of their virtues while minimising to some extent the effects of their deficiencies. This process of optimisation can release a designer from the constraints associated with the selection and manufacture of conventional materials. He can make use of tougher and lighter materials, with properties that can be tailored to suit particular design requirements. And because of the ease with which complex shapes can be manufactured, the complete rethinking of an established design in terms of composites can often lead to both cheaper and better solutions.

The ‘composites’ concept is not a human invention. Wood is a natural composite material consisting of one species of polymer — cellulose fibres with good strength and stiffness — in a resinous matrix of another polymer, the polysaccharide lignin. Nature makes a much better job of design and manufacture than we do, although man was able to recognize that the way of overcoming two major disadvantages of natural wood — that of size (a tree has a limited transverse dimension), and that of anisotropy (properties are markedly different in the axial and radial directions) — was to make the composite material that we call plywood. Bone, teeth and mollusc shells are other natural composites, combining hard ceramic reinforcing phases in natural organic polymer matrices.

A structural composite is a material that consists of two or more phases on a macroscopic scale, whose mechanical performance and properties are designed to be superior to those of the constituent’s materials acting independently. One of the phases is usually discontinuous, stiffer and stronger and is called the *reinforcement*, whereas the less stiff and weaker phase is continuous and is called the *matrix*. The properties of a composite material depend on the properties of the constituents, geometry and distribution of the phases. One of the most important parameters is the volume (or weight) fraction of reinforcement, or fiber volume ratio. The distribution of the reinforcement determines the homogeneity or uniformity of the material system. The more non-uniform is the reinforcement distribution, the more heterogeneous is the material and the higher is the probability of failure in the weakest areas. The geometry and orientation of the reinforcement affect the anisotropy of the system.

The phases of the composite system have different roles that depend on the type and application of the composite material. In the case of low to medium performance composite materials, the reinforcement, usually in the form of short fibres and particles, provides some stiffening but only local strengthening of the material. The matrix, on the other hand, is the main load-bearing constituent governing the mechanical properties of the material. In the case of high performance structural composites, the usually continous-fiber reinforcement is the backbone of the material that determines its stiffness and strength in the direction of fibres. The matrix phase provides protection and support for the sensitive fibres and local stress transfer from one fiber to another. The interphase although small in size, can play an important role in controlling the failure mechanisms, fracture toughness, and overall stress-strain behaviour of the material.

Classification of composites:-

Broadly composites are classified according to the type of matrix or according to the geometry of reinforcement. On the basis of matrix material, composites are classified as: **polymer, metal, ceramic and carbon matrix composites**.

Polymer matrix composites include thermo set (epoxy, polyimide, polyester) or thermoplastic (poly-ether-ether-ketone, polysulfone) resins reinforced with glass, carbon (graphite),aramid(Kevlar) or boron fibers. They are used for relatively low temperature applications**. Metal matrix composites** consist of metals or alloys (aluminium, magnesium, and titanium, copper) reinforced with boron, carbon or ceramic fibers. Their maximum use temperature is limited by softening or melting temperature of the metal matrix. **Ceramic matrix composites** consist of ceramic matrices (silicon carbide, aluminium oxide, glass-ceramic, silicon nitride) reinforced with ceramic fibers. They are best suited for high temperature applications**. Carbon composites** consist of carbon or graphite matrix reinforced with graphite yarn or fabric. They have relatively high strength at high temperatures coupled with low thermal expansion and low density.

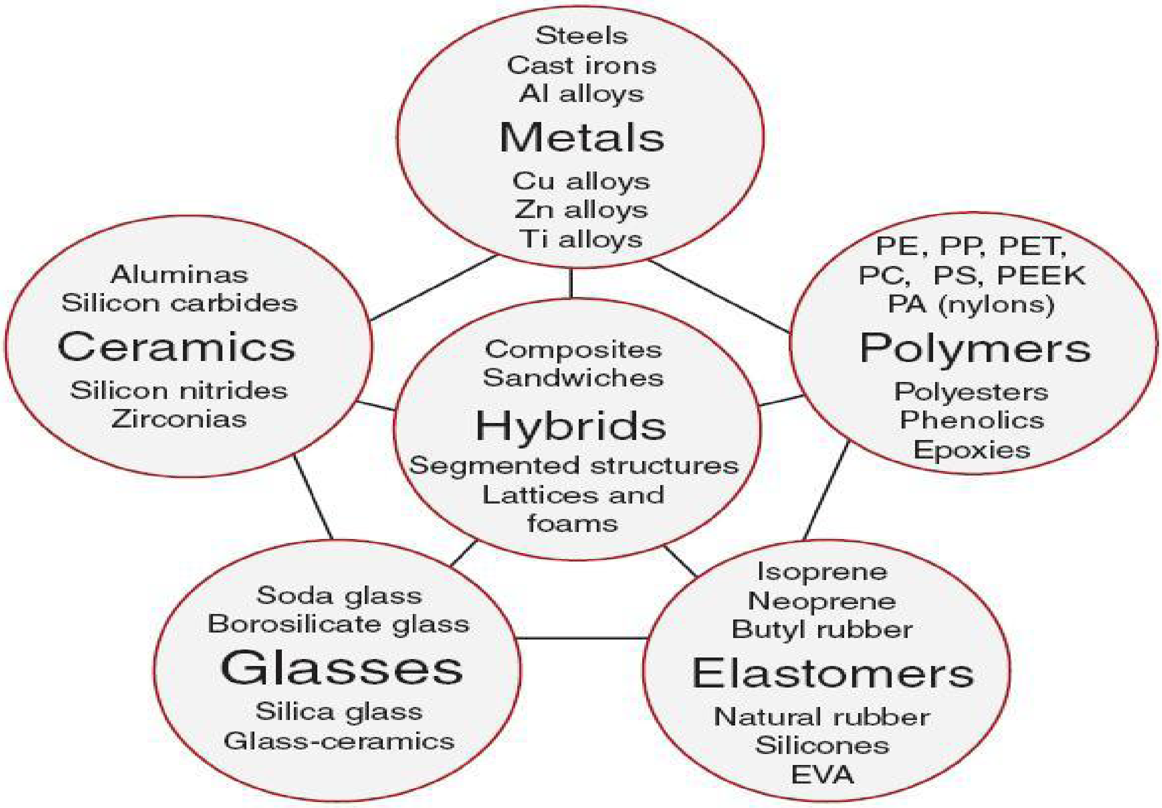


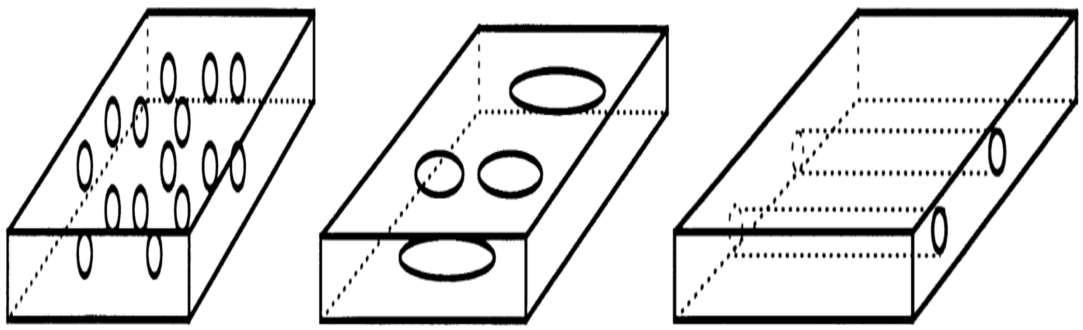
Fig.1.1: Conventional classification of composite materials in accordance to matrix material

On the basis of ***the geometry of reinforcement***, composites are classified as follows:

• ***Particulate*** composites consist of particles of various sizes and sizes randomly dispersed within the matrix. Because of the randomness of particle distribution, these composites can be regarded as quasi homogenous on a scale larger than particle size and spacing and quasi-isotropic. Particulate composites may consist of non-metallic matrix (concrete, glass reinforced with mica flakes, brittle polymers reinforced with rubber like particles); metallic particles in non-metallic matrices (aluminium particles in polyurethane rubber used in rocket propellants); metallic particles in metallic matrices (lead particles in copper alloys to improve mach inability).

• ***Flake***composites consist of flat reinforcements of matrices. Typical flake materials are glass, mica, aluminium, and silver. Flake composites are advantageous due to high out-of-plane flexural modulus, higher strength, and low cost. As flakes cannot be oriented easily, only a limited number of materials are available for use.

• ***Fiber***composites (discontinuous) contain shortfibers or whiskers as the reinforcing phase. by short. In the first instance the composite material tends to be markedly anisotropic and examples include carbon and aramids.The continuous fiber matrix composite are unidirectional, can be oriented at right angles to each other (cross ply or woven fabric continuous fiber composite) or can be oriented along several directions (multidirectional continuous fiber composite).



(Particulate composites) (Flake composites) (Fiber composites)

Fig.1.2: Types of composites based on reinforcement shape

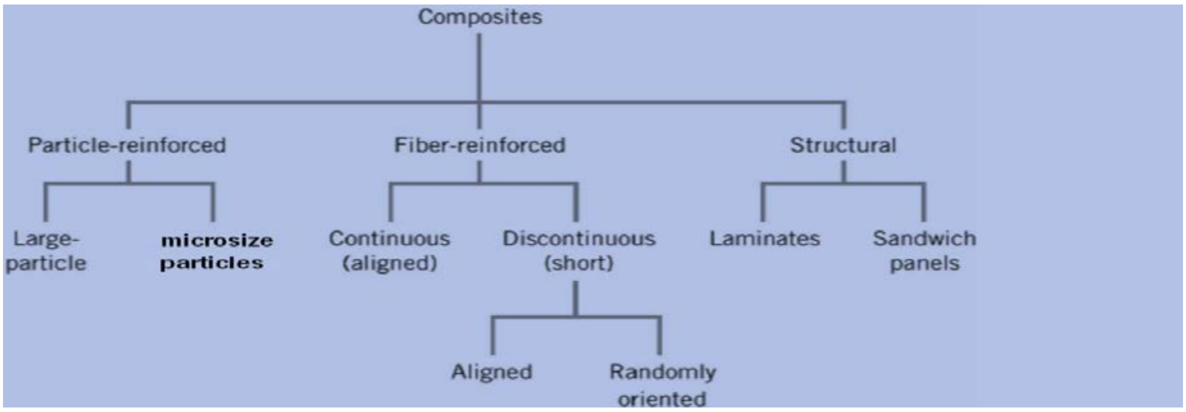


Fig 1.3: Classification of Polymer Composites based on reinforcement type

Fig 1.2 and fig. 1.3 shows the type of composites based on geometry and size of reinforcement respectively.

In the present work, PMCs composites (matrix) are used due their low cost and high strength whereas particulate composites (reinforcement) are preferred as they are isotropic, have increased operating temperature and oxidation resistance.

Advantages of Composite Materials:-

{1}Micromechanics,

{2}Macro mechanics,

{3}Mechanical Characterization,

{4}Structural Design and Optimization,

{5}Fabrication Technology,

{6}Maintainability, Serviceability and Durability

{7}Cost Effectiveness.

Significance of Composite Materials:-

The Studyof composites is a philosophy of material design that allows for the optimum material composition along with structural design and optimization in one concurrent and interactive process. The scope of composite materials research and technology consists of following tasks:

*Investigation of basic characteristics of the constituent and composite materials.

*Material optimization for given service conditions.

*Development of effective and efficient fabrication procedures and understanding of their effect on material properties.

*Development of analytical procedures for determining material properties and prediction of structural behaviour.

*Development of effective experimental methods for material characterization, stress analysis, and failure analysis.

*Non-destructive evaluation of material integrity and structural reliability.

*Assessment of durability, flaw criticality and life prediction.

Application of Composite in Various Fields:-

This is a brief listing of current and proposed applications of composite materials in variousbranchesof industry. It is not intended to be comprehensive or all-embracing, but merely to give an indication of the range of possibilities for designers.

Aerospace

A wide range of load-bearing and non-load-bearing components are already in use in both fixed-wing and rotary wing aircraft. Many military and civil aircraft now contain substantial quantities of lightweight, high-strength carbon-, Kevlar- and glass-fibre composites, as laminated panels and mouldings, and as composite honeycomb structures with metallic or resin-impregnated paper honeycomb core materials. They are used in air frames, wing spars, spoilers, tail-plane structures, fuel tanks, drop tanks, bulkheads, flooring, helicopter rotor blades, propellers, and structural components, pressured gas containers, and landing gear doors, fairings, engine nacelles (particularly where containment capability is required for jet engines), air distribution ducts, seat components, access panels, and so forth. Many modern light aircraft are being increasingly designed to contain as much lightweight composite material as possible. For elevated-temperature applications carbon-fibre-reinforced carbon is in use. Concord's disk brakes use this material, rocket nozzles and re-entry shields have been fashioned from it, and there are other possibilities for its use as static components in jet engines. Rocket motor casings and rocket launchers are also frequently made of reinforced plastics. A particularly interesting (and important) application of composites is in its development in Australia as a means of repairing battle damage (patching) in metal aircraft structures. Space applications offer many opportunities for employing light-weight, high-rigidity structures for structural purposes. Many of the requirements are the same as those for aeronautical structures, since there is a need to have low weight and high stiffness in order to minimize loads and avoid the occurrence of buckling frequencies. Dimensional stability is at a premium, for stable antennae and optical platforms, for example, and materials need to be transparent to radio-frequency waves and stable towards both u-vradiation and moisture.

Automotive Engineering

There is increasing interest in weight reduction in order to permit both energy conservation and increased motoring economy. Reduction in the weight of an automobile structure achieves primary weight-saving and if carried to sufficiently great lengths enables the designer to use smaller power plants, thus achieving substantial secondary improvements in fuel economy. The majority of automotive applications involve glass-reinforced plastics because the extra cost of carbon or aramid fibre is rarely considered to be acceptable in this market. Even so, the cost of using GRP is usually being weighed against the much lower cost of pressed steel components, and the substitution is often rejected on purely economic grounds, leaving aside the question of energy saving. A wide range of car and truck body mouldings, panels and doors is currently in service, including complete front-end mouldings, fascias, bumper mouldings, and various kinds of trim. There is considerable interest in the use of controlledcrushcomponents based on the high energy-absorbing qualities of materials like GRP. Leaf and coil springs and truck drive shafts are also in service, and GRP wheel rims and inlet manifolds have been described in the literature. Selective reinforcement of aluminium alloy components, such as pistons and connecting rods, with alumina fibres is much discussed with reference to increased temperature capability.

Bio-Engineering

Carbon-fibre-reinforced plastic and carbon components are in use for prosthetic purposes, such as inorthopaedic fracture fixation plates, femoral stems for hip replacements, mandibular and maxillaryprostheses (jaw remodelling, for example), and for external orthotic supports in cases of limb deformity*etc*. Pyrolytic carbon is used to manufacture heart valve components, and the substitution of a carbon/carbon composite is not unlikely. There have also been developments in the use of particulate hydroxyapatite as filler in a thermoplastic composite for bone remodelling or replacement.

Chemical Engineering

A substantial amount of GRP is currently in use in chemical plant for containers, pressure vessels, pipe-work, valves, centrifuges etc.

These may be filament-wound or moulded components for containment of process fluids.

Civil/Structural Engineering

Again the bulk of composites used in this field are glass-reinforced plastics. The low inherent elastic modulus of GRP is easily overcome in buildings by the use of double curvature and folded-plate structures: thin GRP panels also offer the advantage of translucency. Glass-reinforced cement (GRC) products made with Cem-FIL (alkali-resistant glass fibres) are gradually being introduced as structural cement-based composites, but these GRC are still regarded with some suspicion by architects who prefer to consider only non-load-bearing applications for glass-reinforced cement. Development of suitable highly-drawn polymer fibres and net-like polymeric reinforcement has made it possible to produce stable polymer-reinforced cement for a variety of purposes. But concrete is the cheapest engineering material available, and it requires very little in the way of expensive reinforcing filaments to be added to it to make it uncompetitive. The answer is usually to use more concrete! But GRC is perhaps likely to attract the more adventurous designer with light-weight concrete structures in mind (thin shell structures for example). A good deal of GRP is used in this industry for folded-plate structures, cladding panels, decorative ‘sculptured’ panels (like those in the doors of the Roman Catholic cathedral in Liverpool), services mouldings and ducting, racking, pipe work, rainwater mouldings, domestic and industrial water tanks, form-work for concrete, and complete small structures like foot-bridges. Light-weight composite panelling for partitioning and similar applications has also been tried. CFRP have been less used until recently because of the cost, but are increasingly being considered for building light-weight structures, including a number of bridges. Current applications include the use of pultruded FRP shapes as individual structural elements and shear stiffeners for concrete structures, as reinforcing bars for concrete, components in composite/concrete structures, as externally applied impact containment supports, and as patches for damaged concrete bridgework.

Domestic

Injection-moulded reinforced thermoplastics and polyester moulding compounds are perhaps the most common composites used in consumer items for the domestic market, and the range is vast. Mouldings of all kinds, from kitchen equipment of all kinds to casings for the whole gamut of domestic and professional electrical equipment, motor-cycle crash helmets, television and computer casings, and furniture.

Electrical Engineering

Typical applications are radomes, structural components for switch gear, power generator coolant containment and large-diameter butterfly valves, high-strength insulators (*eg.* for overhead conductor systems), printed circuit boards, and casings for electronic equipment. The majority of applications in this field again use GRP, although the use of composites which are more thermally stable and more moisture-resistant is increasingly predicated for sensitive, small-scale electronic components. Many prototype and practical wind-generator designs incorporate GRP or hybrid blading.

Marine Engineering

Marine applications include surface vessels, offshore structures and underwater applications. A vast range of pleasure craft has long been produced in GRP, but much serious use is also made of the same materials for hull and superstructure construction of passenger transport vessels, fishing boats and military (mine-countermeasures) vessels. Sea-water cooling circuits may also be made of GRP as well as hulls and other structures. Off-shore structures such as oil rigs also make use of reinforced plastics, especially if they can be shown to improve on the safety of steel structures, for fire protection piping circuits, walkways, flooring, ladders, tanks and storage vessels, blast panels, and accommodation modules. High specific compression properties also make composite materials attractive for submersibles and submarine structures, both for oil exploration and for military purposes, and for towed transducer arrays for sea-bed sonar mapping.

Sport

Perhaps the most visible development in the use of composites has been in the sports goods industry. Manufacturers have been quick to seize on the potential advantages of new materials like carbon and boron fibre composites over conventional wood and metal for sports equipment of all kinds, but whether the average sportsman (and perhaps even some of the above-average ones) who have been inveigled into buying this more expensive composite equipment in the hope that it would improve their game have been able to demonstrate genuine improvement remains uncertain. GRP vaulting poles were perhaps the earliest of the composite sports gear, but one can now obtain tennis rackets, cricket bats, golf clubs, fishing rods, boats, oars, archery equipment, canoes and canoeing gear, surf boards, wind-surfers, skateboards, skis, ski-poles, bicycles, and protective equipment of many sorts in composite materials of one kind or another. In an industry that is often less directly subject to controls exercised in other areas of engineering there is often a tendency to dupe customers with the use of names which incorporate the words ‘carbon’ or ‘graphite’ to describe expensive, black coloured items which may at the present time legitimately contain little or no carbon fibre.

The technology of composite materials has experienced a rapid development in last two decades due to (a) significant progress in materials science and technology in the area of fibers, polymers and ceramics,(b)requirements for high performance materials in aircraft and aerospace structures,(c)development of powerful and sophisticated analysis using modern numerical methods for structural analysis using modern computer technology. Prospects for the future are bright as cost is decreasing due to market expansion, fabrication process is becoming less costly, automation is introduced and availability of many good interactive computer programs for numerical analysis.

Recent electronic devices are packaged with different polymeric materials and usually epoxy or polyster resins are used for encapsulation [1].But due to their low thermal conductivity and high CTE has become a secondary choice for such applications[2].Use of polymers in high cost embedded heat sinks requires new class of particulate filled composites to enhance the thermal conductivity of polymers[3].Improved thermal conductivity in polymers may be achieved by molecular orientation or by the addition of conductive fillers.

Apart, a low dielectric constant is required for fast signal propagation and a low CTE to avoid thermal fatigue [4].Aluminium oxide with a high thermal conductivity, low dielectric constant and low CTE therefore emerges as a suitable filler material to be used in polymeric materials. In view of this, in the present work, aluminium oxide (Al_2O_3) is chosen as the ceramic filler to be dispersed within epoxy resin. The objective of this paper is to analyse the heat transport through the Al_2O_3-epoxy composites and to evaluate the effective thermal conductivity of these composites by numerical as well as experimental method. Through addition of highly conductive inorganic filler into the resin matrix, it is possible to achieve a better thermal conduction path and decrease the thermal contact resistance at the filler–resin matrix interface that attributes to increase in thermal conductivity.

Against this background the present work is focussed on fabrication of a series of such particulate filled composites (epoxy + Al_2O_3) with proper evaluation inthermal conductivity and their characteristics.

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CHAPTER 2

LITERATURE REVIEW

Since 1930's, polymers have made significant advances in the markets of metals, wood, glass, paper, leather, and vulcanized rubber that were conventionally used in most household goods and industrial components as well as creating new markets of their own. The main reason behind the widespread use of polymers is their unique set of properties such as toughness, light weight, low cost, and ease of processing and fabrication. Even though polymers are not the panacea of industry's material problems, their unique set of properties have made them one of the important classes of materials finding their way into widespread use in the electronic industries. The goals for multifunctional electronic packaging materials are that they should be lightweight, injection mouldable into complex shapes, and should have tailored electrical conductivity. Polymers have properties that may help in achieving these goals; however, they are poor thermal conductors, which is the required fundamental property for electronic packaging application. One of the common methods to fabricate materials with multifunctional properties is by having a composite of two or more materials. For the electronic packaging application, a polymer matrix may be filled with conductive fillers to obtain all the above mentioned multifunctional properties. A continuous conductive network of particles throughout the matrix material can improve the effective thermal conductivity of a polymer composite while keeping the intrinsic properties of a polymer.

Thermally conductive polymer composites can replace metals in many applications. This technology is a substantial improvement since polymers are commonly used due to their thermal isolating properties. The advantages of thermally conductive polymers over metals are reduced density; increased corrosion, oxidation, and chemical resistance; increased processibility; and properties are adjustable to fit the application. However, polymers have many disadvantages; for example, creep, thermal instability, and a limited number of processing techniques. The main application for thermally conductive polymers is heat sinks. Other possible benefits are faster injection moulding cycle times and improved thermal stability. The increasing demand for smaller, lighter, and faster machines and electronics has created a need for new materials. In addition, industry has a growing need to tailor the properties of materials, including thermal conductivity, to desired applications.

This literature review provides background information on the issues to be considered in this thesis and emphasizes the relevance of the present study. This treatise embraces some related aspects of polymer composites with special reference to their thermal characteristics. The topics include brief review:

- On Thermal Conductivity of Polymer Composites
- On Particulate Reinforced Polymer Composites
- On Thermal Conductivity Models
- On Polymer Matrix Composites reinforced with Al_2O_3

On Thermal Conductivity of Polymer Composites:-

Different kinds of polymers, and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as heaters, electrodes Jung-il et al [5] , composites with thermal durability at high temperature Nikkeshi et al [6] etc. A few studies have been reported on the thermal conductivity of some filled polymer composites [7-17].WhileKumlutas and Tavman [11] carried out a numerical and experimental study on thermal conductivity of particle filled polymer composites.Tekce et al. [17] noticed the strong influence of the shape factor of fillers on thermal conductivity of the composite. Progellhof.et al. [18] was the first to propose an exhaustive overview on models and methods for predicting the thermal conductivity of composite systems. Mamunya et al. [19] also reported the improvement in electrical and thermal conductivity of polymers filled with metal powders. To estimate the effective thermal conductivity of fibrous reinforced composite materials a numerical approach, using a finite-element formulation carried out on a three-dimensional geometric space was proposed by Veyret et al. [12, 43]. Procter and Solc[20] used Nielsen model as a prediction to investigate the thermal conductivity of several types of polymer composites filled with different fillers and confirmed its applicability. Nagai and Lai [21] found that Bruggeman model for Al_2O_3/epoxy system and a modified form of Bruggeman model for AlN/epoxy system are both good prediction theories for thermal conductivity. In a recent research Weidenfeller et al. [22] studied the effect of the interconnectivity of the filler particles and its important role in the thermal conductivity of the composites.

On Particle Reinforced Polymer Composites:-

Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications while hard particulate fillers consisting of ceramic / metal particles and fiber fillers made of glass are extensively being used to improve the mechanical properties such as wear resistance [23-25]. Relative case of processibility, low density, high corrosion resistance and low cost in these composites has driven them as potential candidates for development of devices for electric stress control and high storage capability and high permittivity material. Ceramic filled polymer composites have been the subject of extensive research in last two decades. The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at the cost reduction and stiffness improvement [26,27].When silica particles are added into a polymer matrix to form a composite, they play an important role in improving electrical, mechanical and thermal properties of the composites [28,29]. Currently, particle size is being reduced rapidly and many studies have focused on how single-particle size affects mechanical properties [30-36]. Yamamoto et al. [37] reported that the structure and shape of silica particle have significant effects on the mechanical properties such as fatigue resistance, tensile and fracture properties. Nakamura et al. [38-40] discussed the effects of size and shape of silica particles on the strength and fracture toughness based on particle-matrix adhesion and also found an increase of the flexural and tensile strength as specific surface area of particles increased.Pattnaiket al.[41] reported the existence of a possible correlation between thermal conductivity and wear resistance of particulate filled composites.Nayak et al. [42] have reported on the modified thermal conductivity of pine wood dust filled epoxy based composites. Hill and Supancic [43] proposed an indirect method to determine this interfacial boundary resistance by preparing large-scale “macro-model” simulations of the polymer-ceramic interface. They also investigated the effects of similar size and shape of plated shaped particles on the thermal conductivity of polymer/ceramic composite materials.

On Polymer Matrix Composites reinforced with Al_2O_3:-

Experience on the use of aluminium oxide as a filler of epoxy matrices is very limited. In the best case, investigations have been performed with micro sized aggregates of nano-particles, because the filler has been added to the resin in the form of a powder [44]. The reinforcement of the epoxy matrix by aluminium oxide particles is especially attractive, because these fillers increase the strength and thermal stability of the material and impart resistance to corrosive media, except for strongly alkaline media.In an attempt to develop a rapid and economical method for making tools with low volume production, some particulate reinforced epoxy composites were investigated by S.Ma et al.[45].The tools built up using this method are suitable for wax and polymer materials injection moulding applications.S.Biswaset. al. [46] describes the development of alumina filled glass fiber- reinforced epoxy matrix composites and compared the experimental results obtained from Taguchi experimental design with the theoretical erosion model. These composites find applications in highly erosive environments.A.Pattnaiket. al [47] studied an abrasive wear behaviour of randomly oriented glass fiber reinforced with epoxy resin filled Al_2O_3, SiC and pine bark dust.Recently,K.Sabeel Ahmed et.al[48] studied the effect of ceramic fillers like SiC and Al_2O_3 on wear behaviour of jute/epoxy composites and observed that Al_2O_3 filled composites has better wear resistant than SiC filled composites. An experimental study is carried out to investigate bearing strength behaviour of pinned joints of glass fiber reinforced composites by O.Asi[49] and hence concluded that bearing strength first increased with 15wt% Al_2O_3 filler content and then decreased with further increase in Al_2O_3 content due to low void content in the previous concentration.Y.Wanget.al[50] studied the tribological and electrochemical corrosion properties of Al_2O_3 /polymer nano-composites coatings and showed improvement in scratch and abrasive resistance for 20 wt% Al_2O_3 compared with that of polymer coating.B.N.Dudkinet.al[51] studied that filling Al_2O_3 nanoparticles in matrices increases the mechanical strength of the composite to a larger extent whereas filling of the matrices with Al_2O_3nanofibers favours an increase in the Young modulus.

There are only a few published papers on evaluation of effective thermal conductivity of epoxy composites filled with alumina and till now no efforts have been made to study the effect of filler content using a numerical approach.L.C.Sim.et al [52] investigated using Al_2O_3 or ZnOfillers, to analyse theeffect of thermal conductivity on thermal pads.D.C.Moreira. et. al[53] conducted experimentation to determine how the effective thermal conductivity is influenced by the addition of alumina (Al_2O_3) and tenorite (CuO) nano-particles to an unsaturated polyester resin (UPR) matrix,which has a lower thermal conductivity compared to the employedmetal oxides.

In view of the above, the present work is undertaken to fabricate and investigate thermally conductive particulate filled polymer composites with maximum possible thermal conductivity and minimum coefficient of thermal expansion.

On Thermal Conductivity Models:-

Many theoretical and empirical models have been proposed to predict the effective thermal conductivity of two-phase mixtures [18, 54]. For a two-component composite, the simplest alternatives would be with the materials arranged in either parallel or series with respect to heat flow, which gives the upper or lower bounds of effective thermal conductivity

Parallel conduction model:

$$k_c = (1-ø)k_m + \phi k_f \tag{2.1}$$

Where k_c=thermal conductivity of composite

K_m =thermal conductivity of matrix

K_f=thermal conductivity of filler

Ø = volume fraction of filler.

Series conduction model:

$$\frac{1}{k_c} = \frac{(1-ø)}{k_m} + \frac{ø}{k_f} \tag{2.2}$$

The equations (1) and (2) are called as the Rules of Mixture (ROM) model.

For two-phase solid mixture, thermal conductivity of the individual components parameters was derived by Tsao[54]. Cheng and Vachon [55] assumed a parabolic distribution of the discontinuous phase in the continuous phase, to obtain a solution to Tsao’s model that did not require knowledge of additional parameters.

According to Agari and Uno [56] model, the expression that governs the thermal conductivity of the composite is:

$\log(k_c) = \o C_2 \log(k_f)+(1-\o)\log(C_1\, k_m)$ (2.3)

Where, C_1, C_2 are experimentally determined constants of order unity.

The derived expression for effective thermal conductivity of dilute composite of spherical particles is

$$\frac{k}{k_c} = 1 + \frac{3(k_d - k_c)}{(k_d + 2k_c)} \tag{2.4}$$

$$k_c = k_m \left[\frac{k_f + 2k_m - 2\o(k_m - k_f)}{k_f + 2k_m + \o(k_m - k_f)} \right] \tag{2.5}$$

Where K=thermal conductivity of composite,

K_c=thermal conductivity of continuous-phase (matrix),

K_d=thermal conductivity of dispersed-phase (filler), and

Ø = volume fraction of the dispersed phase.

Equation (4) and (5) is the well-known Maxwell equation [57] for dilute composites.

Lewis and Nielsen [58] modified the Halpin–Tsai equation [59-61] which includes effect of shape of the particles and orientation or type of packing for a two-phase system:

$$k_c = k_m \left[\frac{1+AB\emptyset}{1-B\emptyset\psi}\right] \tag{2.6}$$

where ,

$$B = \frac{\frac{k_f}{k_m} - 1}{\frac{k_f}{k_m} + A} \quad , \quad \psi = 1 + \left(\frac{1-\phi_m}{{\phi_m}^2}\right)\phi$$

Table.2.1 Value of *A* for various systems [11]

Type of dispersed phase	Direction of heat flow	*A*
Cubes	Any	2
Spheres	Any	1.5
Aggregates of spheres	Any	(2.5/ $Ø_m$) - 1
Randomly oriented rods Aspect ratio=2	Any	1.58
Randomly oriented rods Aspect ratio=4	Any	2.08
Randomly oriented rods Aspect ratio=6	Any	2.8
Randomly oriented rods Aspect ratio=10	Any	4.93
Randomly oriented rods Aspect ratio=15	Any	8.38
Uniaxially oriented fibers	Parallel to fibers	2L/D
Uniaxially oriented fibers	Perpendicular to fibers	0.5

TABLE 2.2: Values of $Ø_m$[11]

Shape of particle	Type of packing	$Ø_m$
Spheres	Hexagonal close	0.7405
Spheres	Face centered cubic	0.7405
Spheres	Body centered cubic	0.60
Spheres	Simple cubic	0.524
Spheres	Random close	0.637
Rods and fibers	Uniaxial hexagonal close	0.907
Rods and fibers	Uniaxial simple cubic	0.785
Rods and fibers	Uniaxial random	0.82
Rods and fibers	Three dimensional random	0.52

The values of 'A' and 'ϕ_m'depend on geometric shapes and orientation.

[Value of A=1.5 and for ϕ_m=0.524.]

J.Z.Lianget. al [62] made efforts to study the heat transfer process and mechanisms in inorganic hollow micro-sphere filled polymer composites and also established relevant mathematical model to calculate effective thermal conductivity for these composites.Recently,Agrawaland Satapathy[63] proposed a theoretical model for single filler to predict the effective thermal conductivity of the particulate filled polymer matrix composites.

Knowledge Gap in Earlier Investigations:-

In the past, though a number of intensive studies had been published on the thermal conductivity of particulate composites there is awide knowledge gap that demands a well-planned and systematic research in this area of particulate filled polymer composites. From review of the published literature reveals that:

- Most of the investigations are aimed at enhancing the thermal conductivity of the polymer rather than attempting to modify both thermal and dielectric properties for micro-electronics applications.
- There is no report available on evaluation of effective thermal conductivity of alumina particulates filled polymer composites.
- Investigation of thermal conductivity ofalumina particulates filled epoxy composite using numerical modelling has not been reported.
- The understanding of the relationship between the effective thermal conductivity of a composite material and the micro-structural properties (volume fractions, distribution of particles, aggregation of particles, properties of individual components, etc.) is far from satisfactory.

A comprehensive and systematic evaluation of thermal behaviour of Al_2O_3 filled epoxy composites has not adequately been performed yet. In view of the above, the present work is undertaken to investigate on the thermal characteristics such as effective thermal conductivity, glass transition temperature, coefficient of thermal expansion etc.

Objectives of Present Work:-

The objectives of this work are outlined as follows:

1. To fabricate a set of epoxy composites with micro sized alumina as filler, with an objective to modify the heat conduction capability needed for micro-electronics applications.
2. Development of numerical models for estimation of K_{eff} of such particulate filled polymer composites.
3. Comparison among the K_{eff}values from proposed numerical model and those obtained from existing correlations.
4. Validation of developed numerical model by measuring effective thermal conductivity values experimentally.
5. Physical Characterization of these fabricated composites and studying the effect of Al_2O_3 content on certain thermalcharacteristics.

Chapter Summary

This chapter provides an outline of research works on particulate reinforced polymer composites, thermal conductivity of polymer matrix composites, thermal conductivity models made by various investigators.

The subsequent chapter discusses the fabrication methods, characterization details and the finite element formulation in brief comparison with experimental results.

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CHAPTER 3

MATERIALS AND METHODS

Materials

(a)Matrix material:

Epoxy resins are the most commonly used resins. Epoxy resins are low molecular weight pre-polymers or higher molecular weight polymers which normally contain at least two epoxide groups. The epoxide group is also sometimes referred to as a glycidyl or oxirane group. A wide range of epoxy resins are produced industrially. The raw materials for epoxy resin production are today largely petroleum derived; although some plant derived sources are now becoming commercially available (e.g. plant derived glycerol used to make epichlorhydrin). Epoxy resins are polymeric or semi-polymeric materials, and as such rarely exist as pure substances, since variable chain length results from the polymerisation reaction used to produce them. High purity grades can be produced for certain applications, e.g. using a distillation purification process. One downside of high purity liquid grades is their tendency to form crystalline solids due to their highly regular structure, which require melting to enable processing. The most common and important class of epoxy resins is formed from reacting epichlorhydrin with bisphenol A to form diglycidyl ethers of bisphenol A. The simplest resin of this class is formed from reacting two moles of epichlorhydrin with one mole of bisphenol A to form the bisphenol-A-diglycidyl ether (commonly abbreviated to DGEBA or BADGE). DGEBA resins are transparent colourless-to-pale-yellow liquids at room temperature, with viscosity typically in the range of 5-15 Pa.s at 25°C. Industrial grades normally contain some distribution of molecular weight, since pure DGEBA shows a strong tendency to form a crystalline solid upon storage at ambient temperature.

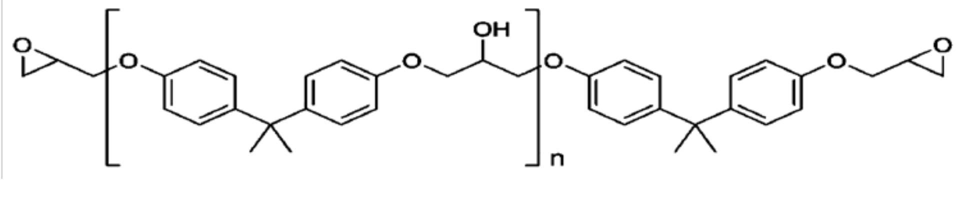

Fig 3.1: Unmodified epoxy pre polymer resin chain.

Epoxy is chosen primarily because it happens to be the most commonly used polymer and because of its insulating nature (low value of thermal conductivity, about 0.363 W/m K). The low temperature curing epoxy resin (Araldite LY 556) and corresponding hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. The epoxy resin and the hardener are supplied by Ciba Geigy India Ltd.

Fig3.2: Epoxy Resin

(b)Filler material (Al_2O_3):-

Aluminium oxide is a chemical compound of aluminium and oxygen with the chemical formula Al_2O_3.It is commonly called **alumina**, and may also be called **aloxide**, **aloxite**, or**alundum** depending on particular forms or applications. Alumina contributes 15% of the earth's crust and is amphoteric in nature. It has strong ionic inter-atomic bonding. It commonly occurs in its crystalline polymorphic phase α-Al_2O_3, in which it comprises the mineral corundum, varieties of which form the precious gems ruby and sapphire. Al_2O_3 is significant in its use to produce aluminium metal, as an abrasive owing to its hardness, and as a refractory material owing to its high melting point.Al_2O_3 is an electrical insulator and has a relatively high thermal conductivity (35 W/m-K).

Key properties: High strength and hardness, high temperature stability, high corrosion resistance, high wear resistance.

Applications: Used in electrical industry for insulation parts, electronics industry as a substrate, protective corrosion coatings and high temperature applications.

Fig. 3.3: Commercially available alumina particles

Table 3.1: Engineering Properties of Aluminum Oxide

Properties	Values	Units
Density	3.89	gm/cc
Flexural Strength	379	MPa
Elastic Modulus	375	GPa
Shear Modulus	152	GPa
Bulk Modulus	228	GPa
Poisson's Ratio	0.22	----
Compressive Strength	2600	MPa
Hardness	1440	Kg/mm^2
Maximum Use Temperature	1750	°C
Thermal Conductivity	**35**	**W/m°K**
Dielectric Constant at 20^0C @ 1 MHz	9.8	--------
Resistivity at 20^0C	$>10^4$	Ohm-cm
Dielectric Rigidity at 50 Hz	30	kV/mm
Coefficient of Thermal Expansion	8.4	10^{-6}/°C

Composite Fabrication:-

Low temperature curing Epoxy LY 556 resin, used as the matrix material and the hardener (HY951) are mixed in a ratio of 10:1 by weight as recommended. Epoxy is chosen primarily of its low density (1.1 gm/cc) and low value of thermal conductivity (0.363 W/m-K). Solid alumina particulates are reinforced in the resin to prepare the composites. The dough (epoxy filled with alumina) is then slowly decanted into disc type cylindrical glass moulds, coated beforehand with a uniform thin film of silicone-releasing agent. The composites are cast in these moulds so as to get cylindrical specimens (dia20 mm, thickness 5 mm). The castings are left to cure at room temperature for about 24 hours to undergo complete polymerization, after which the glass moulds are broken and samples are released for further physical characterization and thermal conductivity test.

Table 3.2: List of particulate filled composites fabricated by hand-lay-up technique (FEM analysis)

Sample	Composition
1	Epoxy +1.4 vol%(4.78 wt%)filler
2	Epoxy +3.35vol%(10.91 wt%)filler
3	Epoxy +5.23 vol%(16.35 wt%)filler
4	Epoxy +7.85vol%(23.15 wt%)filler
5	Epoxy +9.42vol%(26.84 wt%)filler
6	Epoxy +11.3vol%(31.05 wt%)filler

Table 3.3: List of particulate filled composites fabricated by hand-lay-up technique (EXPERIMENTAL ANALYSIS)

Sample	Composition
1	Epoxy +1.4 vol%(4.784 wt%)filler
2	Epoxy +2.75 vol%(9.085 wt%)filler
3	Epoxy +5.35 vol%(16.66 wt%)filler
4	Epoxy +6.6 vol%(20 wt%)filler
5	Epoxy +7.82 vol%(23.08 wt%)filler
6	Epoxy +10.8 vol%(29.98 wt%)filler
7	Epoxy +13.2 vol%(34.56 wt%)filler
8	Epoxy +15.9 vol%(40.07 wt%)filler
9	Epoxy +18.8 vol%(45.02 wt%)filler
10	Epoxy +22.1 vol%(50.08 wt%)filler

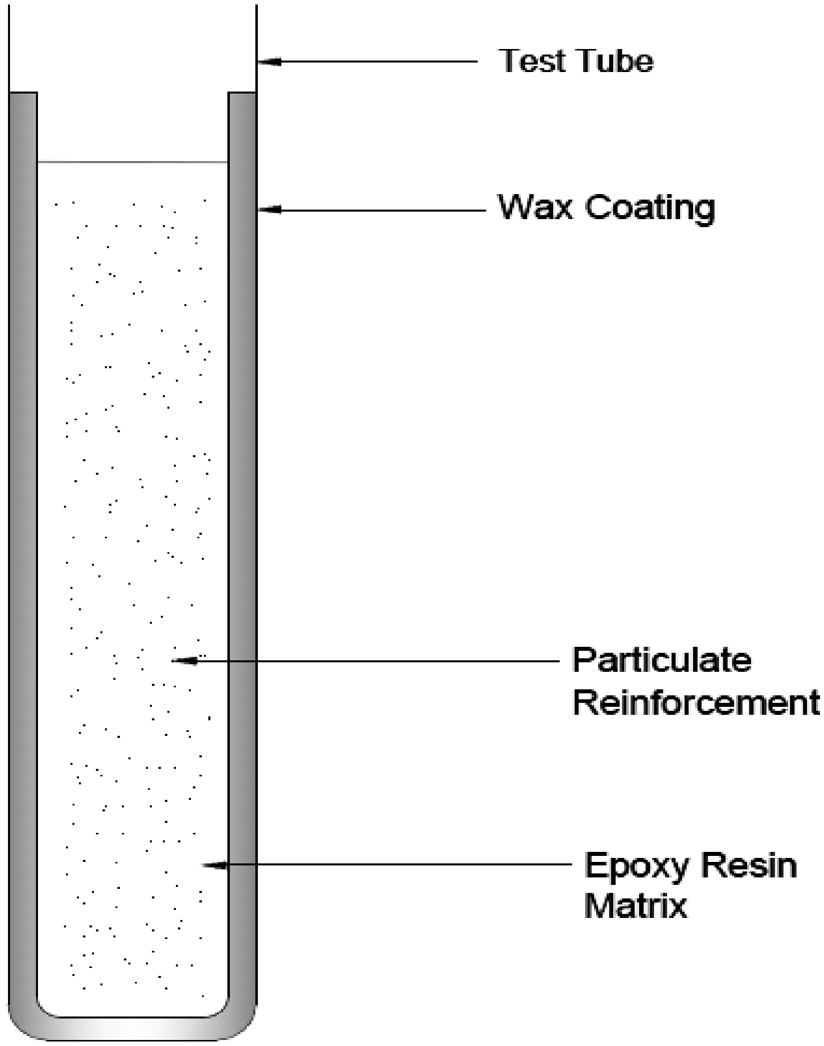

Fig 3.4: Preparation of Epoxy filled with Alumina composite

Fig.3.5: Samples of particulate filled composites fabricated by hand-lay-up technique

Experimental Determination of Thermal Conductivity:-

The Unitherm^{TM} Model 2022(Guarded heat flow meter Thermal Conductivity system) is used to measure thermal conductivity of epoxy filled with alumina particulate composites. Only a relatively small test sample is required. The tests are in accordance with ASTM E-1530Standard. The device has an airtight compartment to keep it moisture free.

Each sample of the prepared composite is kept under a uniform compressive load between two polished surfaces having different temperatures. The lower surface is part of a calibrated heat flow transducer while from the upper surface heat flows into the sample developing an axial temperature difference in the stack. When thermal equilibrium is reached, ΔT across sample is recorded by temperature sensors along with the output from the heat flow transducer. The data recorded and sample thickness is evaluated to calculate the thermal conductivity.

Fig 3.6: Determination of Thermal Conductivity using Unitherm™ Model 2022

To reduce heat transfer across edges, test stack is surrounded by guardfurnace.Around 45 to 60 mins.is needed to conduct the test. It is unavoidable to have a substantial temperature difference between the cold face of the sample and the heat sink and hence, a city water cooled heat sink is provided (about 50^{o}C).

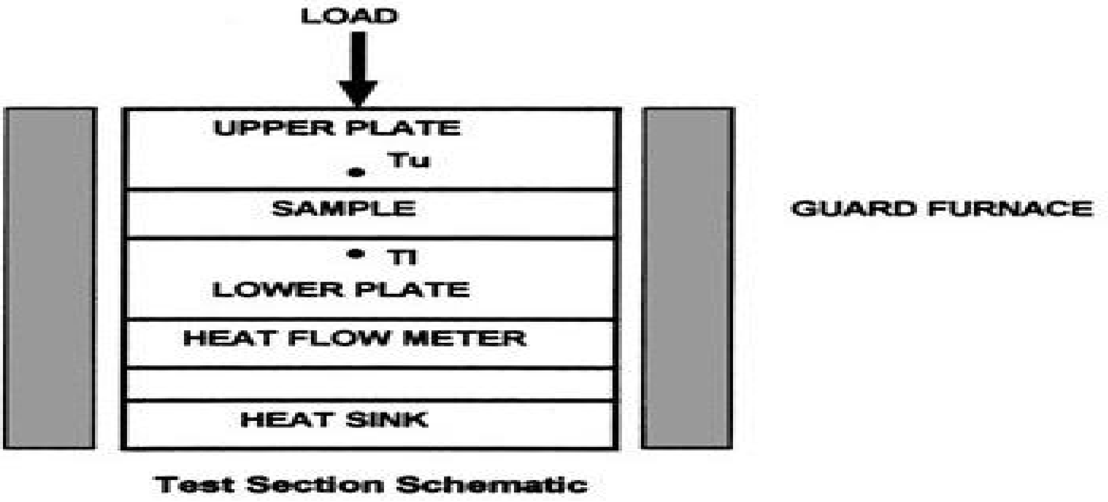

Fig.3.7: Schematic model showing the system arrangement in Unitherm 2022

For one dimensional heat conduction, Q=KA(T_1-T_2)/X ------------[3.1]

and thermal resistance of the sample is given by R=(T_1-T_2)Q/A ----------[3.2],where R is the resistance of the sample between hot and cold surfaces(m^2 K/W).

From Eq(3.1) and Eq(3.2) we can derive that K=x/A --------------[3.3]

In Unitherm Model2022, the heat flux transducer measures the Q value and ΔTbetween both the plates is obtained. Now thermal resistance of sample is evaluated between the upper and lower surfaces. Substituting sample thickness and known cross-sectional area, the thermal conductivity of the samples can be calculated using Eq(3.3).

Thermal mechanical analysis

The glass transition temperature (*Tg*) were measured with a Perkin Elmer DSC-7 thermal mechanical analyzer (TMA). The temperature range used was from 30 to 220^{0} C, and the heating rate was 10^{0}C/ min. All reported DSC/TMA data are obtained from a second heating cycle.

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CHAPTER 4

PHYSICAL CHARACTERIZATION OF EPOXY/Al_2O_3 COMPOSITES

Density and void fraction:

The theoretical density of composite materials in terms of weight fraction can easily be obtained as for the following equations given by Agarwaland Broutman [64].

$$\rho_{ct} = 1/ \{w_m/\rho_m + w_f/ \rho_f\} \qquad (4.1)$$

$$w_f = \rho_f * v_f/ \{ \rho_f * v_f + \rho_m * v_m\} \qquad (4.2)$$

Where, *W* and *ρ* represent the weight fraction and density respectively. The suffix *f*, *m* and *ct* stand for the fiber, matrix and the composite materials respectively.

In case of hybrid composites, consisting of three components namely matrix, fiber and particulate filler, the modified form of the expression for the density of the composite can be written as:

$$\rho_{ct} = 1/ \{w_m/\rho_m + w_f/ \rho_f + w_p/ \rho_p \} \qquad (4.3)$$

Where the suffix *p'*indicates the particulate filler materials.

Table 4.1: Density of Composites

SL NO.	VOLUME FRACTION (%)	WEIGHT FRACTION (%)	THEORETCAL DENSITY	MEASURED DENSITY	POROSITY (%)
1	2.5	8.289	1.169	1.156	1.112
2	5	15.64	1.239	1.226	1.049
3	7.5	22.248	1.309	1.292	1.299
4	10	28.128	1.379	1.361	1.305
5	12.5	33.224	1.445	1.41	2.422
6	15	38.233	1.516	1.48	2.375
7	17.5	42.801	1.588	1.554	2.141
8	20	46.86	1.658	1.618	2.413
9	22.5	50.693	1.728	1.676	3.009
10	25	54.524	1.797	1.732	3.617

The actual density ($\dot{\rho}_{cc}$)of the composite, however, can be determined experimentally by simple water immersion technique. The volume fraction of voids/porosity in the composites is calculated using the following equation:

$$\text{Porosity/voids} = \{\rho_{ct} - \rho_{cc}\}/\rho_{ct} \qquad (4.4)$$

The theoretical and measured densities along with the corresponding volume fraction of voids were presented in Table 4.1. It may be noted that the composite density values calculated theoretically from weight fractions using Eq(4.1) were not in agreement with the experimentally determined values. The difference was a measure of voids and pores present in the composites. It was clear from the Table 4.1 that with the addition of filler materials more voids were found in the composites. As the filler content changes from composites to composites the volume fraction of voids was also found to be change.

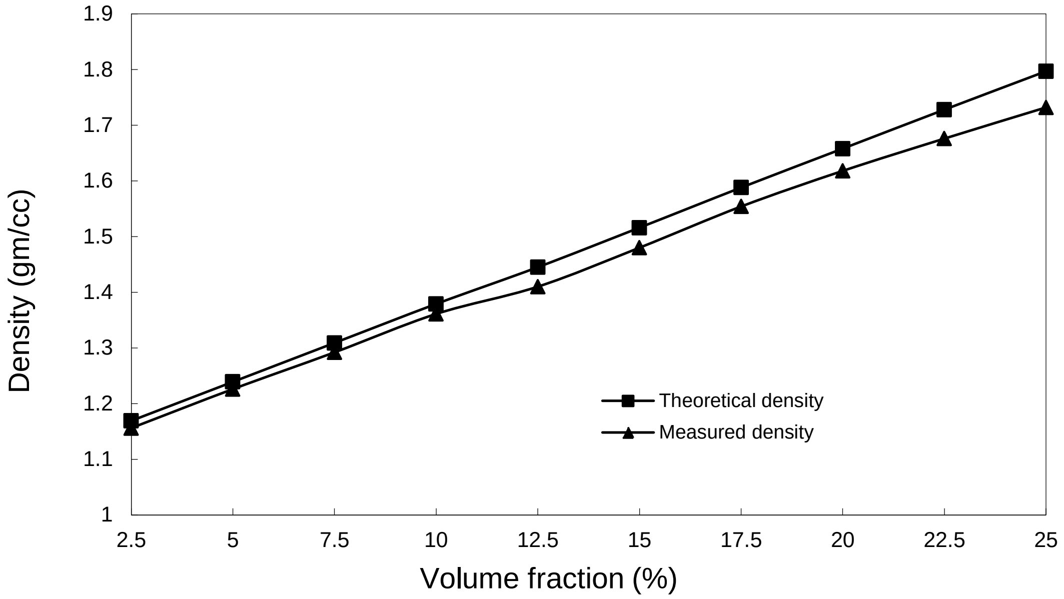

Fig 4.1: Measured and Theoretical Densities for Al_2O_3/epoxy composites.

A comparison of measured and theoretical density values is represented on fig 4.1 for Al_2O_3/epoxy composites with different filler volume fractions. Density of a composite depends on the relative proportion of matrix and reinforcing materials and this was one of the most important factors determining the properties of the composites. The void/porosity content was the cause for the difference between the values of true density and the theoretically calculated one. The voids significantly affect some of the mechanical properties and even the performance of composites in the place of use. Higher void contents usually mean lower fatigue resistance, greater susceptibility to water penetration and weathering. The knowledge of void content was desirable for estimation of the quality of the composites. It was understandable that a good composite should have fewer voids. However, presence of void was unavoidable in composite making particularly through hand-lay-up route.

SEM Analysis:-

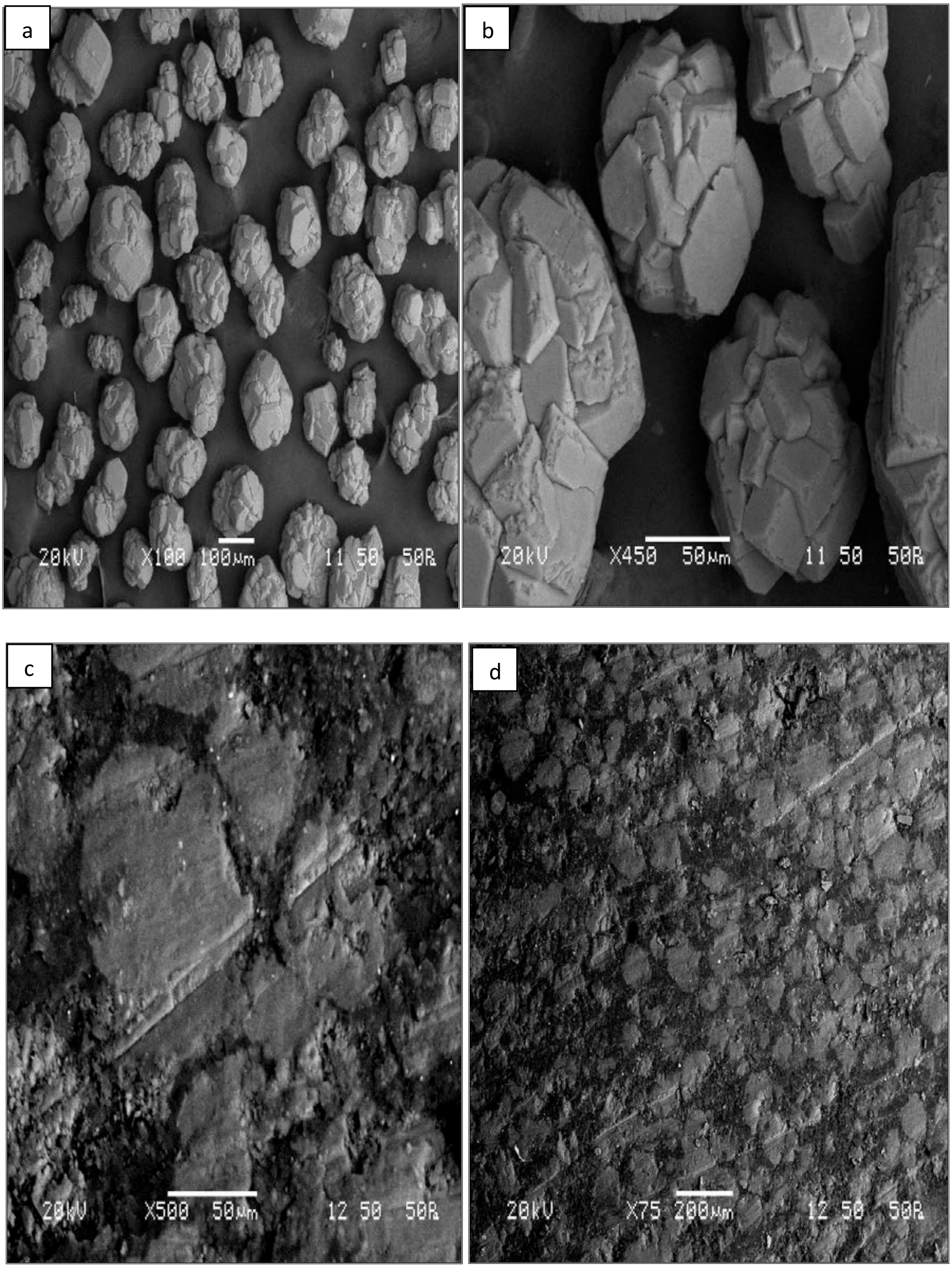

Fig.4.2 (a)-(d): Typical SEM images of fracture surface of the composite

It is well known that the properties of the composites are strongly dependent on the interaction of the filler and polymer. In order to evaluate this interaction, the microstructure of the composites, including the dispersion of Al_2O_3 fillers into epoxy matrix was observed by SEM. Fig. 4.2 shows the typical SEM images. The microstructures reflect particle dispersion in the entire epoxy matrix composite.Fig.4.2a and b shows lots of Al_2O_3microparticles having spherical geometry. This has been clearly observed when the magnification of the images was improved. Furthermore, the interlayers of composites are clearly depicted in fig.4.2c and d indicating that the epoxy molecules were strongly bonded with Al_2O_3 micro balls and a few gaps were observed around these micro balls. With an increase in the Al_2O_3 content, denser composites were obtained. Such mutual contact of Al_2O_3 particles forms a thermal conductive pathway that heightens the thermal conductivity of epoxy/Al_2O_3 composites.WithAl_2O_3particlesdispersed uniformly in the whole epoxy matrix, resembles good homogeneity of the mechanical properties of the composites.

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CHAPTER 5

RESULTS AND DISCUSSION

To predict the effective thermal conductivity of composite materials, several theoretical and empirical models have been proposed in the past. Thermal conductivities for low filler concentrations are predicted very well using this models but when there is an increase in filler concentrations, conductive chains arises and particle agglomeration becomes large which makes difficult to evaluate effective thermal conductivity using above model.

The theoretical analysis of heat transfer in composite material is based on the following assumption:

(a) Locally both the matrix and fillers are homogeneous and isotropic.
(b) The thermal contact resistance between the fillers and the matrix is negligible and the composite lamina is free from voids.
(c) The temperature distribution along the direction of heat flow is linear.

A theoretical model [63] for one dimensional heat conduction through such a composite system has been developed using the law of minimal thermal resistance and equal law of the specific equivalent thermal by putting the expression of all the thermal resistance into the effective thermal conductivity model and deduced as

$$k_{eff} = \frac{1}{\frac{1}{k_p} - \frac{1}{k_p}\left(\frac{6\phi_f}{\pi}\right)^{\frac{1}{3}} + \frac{4}{\left(k_p\left(\frac{4\pi}{3\phi_f}\right)^{\frac{2}{3}} + \left(\frac{2\phi_f}{9\pi}\right)^{\frac{1}{3}} 2\pi\left(k_f - k_p\right)\right)}} \tag{5.1}$$

WhereΦ_f represents the volume fraction ,k_f is the thermal conductivity of filler, k_p is the thermal conductivity of polymer matrix.

PART A: Experimental Determination of K_{eff} and comparison with Theoretical value

The effective thermal conductivity of filled polymer composites depends not only on the component properties and the filler content, but also on the filler shape, filler distribution, and interaction between the filler particles.The phenomenon involving the change in the dispersion state of the conducting phase is known as percolation. Percolation deals with the effects of varying the connectivity of elements (e.g., particles, sites, or bonds) on random system i.e. long-range connectivity. According to the experimentally determined effective thermal conductivity of composites, the K_{eff}increases rapidly when the filler volume fraction reaches 16-22vol% as shown in the fig 5.1. Furthermore, the effective thermal conductivity is reinforced nonlinearly with the increase of the filler content due to the gradual development of the density of the network. Below the percolation threshold, the conductivity is negligible and the threshold conductivity of the composites is equal to the polymer conductivity or slightly higher.

Table5.1: Determination of experimental values (K_{eff}) for different filler concentrations

SL NO	VOLUME FRACTION (%)	EXERIMENTAL VALUE (W/m-K)	THEORETICAL VALUE (W/m-K)
1	1.4	0.449	0.493
2	2.75	0.492	0.549
3	5.35	0.588	0.643
4	6.6	0.612	0.686
5	7.82	0.639	0.727
6	10.8	0.740	0.832
7	13.2	0.836	0.922
8	15.9	0.927	1.029
9	18.8	1.815	1.159
10	22.1	1.973	1.332

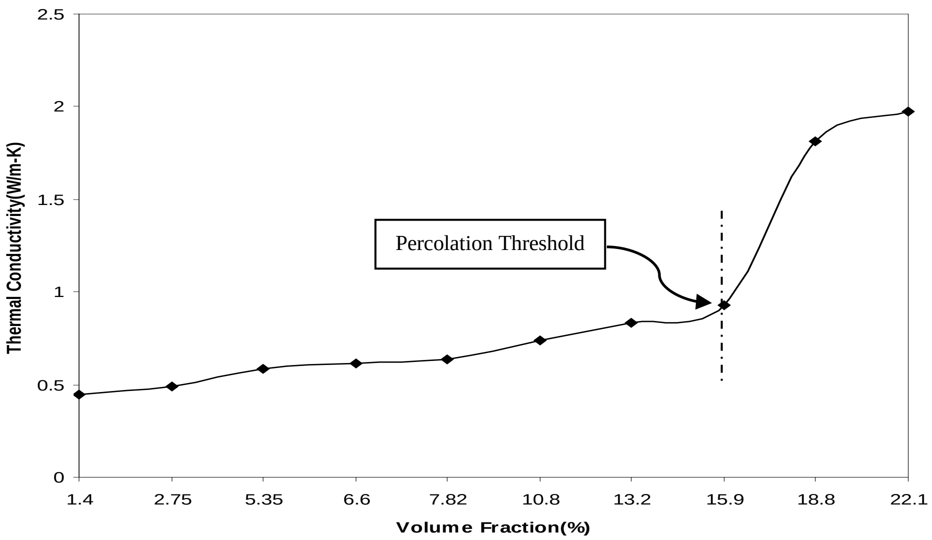

Fig.5.1: Thermal Conductivity of Al_2O_3 filled epoxy composites

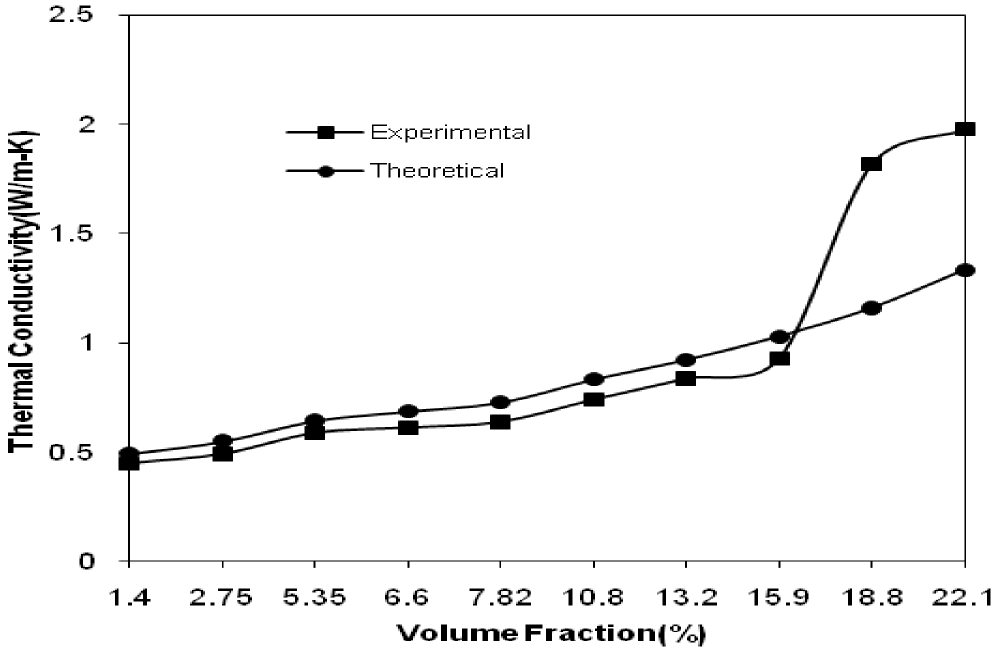

Fig5.2: Comparison of K_{eff} values for Al_2O_3 filled epoxy composites

Using the deduced correlation given by Eqn. (5.1), the effective thermal conductivities (K_{eff}) of all the composites are calculated. Fig.5.2 also presents a comparison of the calculated values with the experimentally measured ones. The 2D and 3D finite element models of filled polymer composites [65-66] cannot reflect the actual microscopic irregularities of the filler shape and distribution, especially for the case of high filler content when a continuous network of filler is formed. It is seen that the results obtained from the proposed correlation are in good agreement with experimental results up to a filler concentration of about 16vol%. The Al_2O_3 particles show a percolation behavior at this volume fraction (16vol %) at which a sudden jump in the thermal conductivity is noticed. This is the critical concentration at which Al_2O_3 particles start contacting with each other and hence the actual size of the agglomerates becomes larger. Consequently, the heat conduction performance of epoxy composites incorporating Al_2O_3 exceeds expectations.

PART B: Numerical Analysis: Concept of Finite Element Method and ANSYS

The finite element method (FEM), originally introduced by Turner et al. [67], is a powerful computational technique for approximate solutions to a variety of ‘‘real-world” engineering problems having complex domains subjected to general boundary conditions.

The Finite Element Analysis (FEA) is a numerical method useful for solving problems with complicated geometries, loadings, and material properties where analytical solutions cannot be obtained. The finite element method (FEM) technique helps to obtain approximate solutions of partial differential equations. One technique is based on eliminating the differential equation completely and work with a minimization problem. Advantages: Flexibility with respect to boundary conditions and geometries and easy to incorporate adequate spatial resolution by varying the element size manually or adaptively.

The goal of numerical simulation is to make predictions concerning the response of physical systems to various kinds of excitation and, based on those predictions, make informed decisions. Therefore mathematical models are defined and finally numerical solutions are calculated. Mathematical models should be understood to be idealized representations of reality and should never be confused with the physical reality that they are supposed to represent. Through conceptualization process a mathematical model is formulated. By discretization process, exact solution of the mathematical model is approximated and by extraction process genuine data's are computed from the approximate solution.

The finite element method (FEM) is a powerful tool used in numerical methods to calculate approximate solutions to mathematical problems so that it can simulate the responses of physical systems to various forms of excitation. It can be widely used in engineering and science, such as elasticity, heat transfer, fluid dynamics, electromagnetism, acoustics, biomechanics, etc. In FEM, the domain is decomposed into a finite number of sub-domains (elements) and exact approximate solution is obtained by the variational or weighted residual methods. ANSYS is general-purpose finite element modelling package used to solve numerous mechanical problems involving static/dynamic, structural analysis (both linear and nonlinear), heat transfer, fluid problems, as well as acoustic and electromagnetic problems. In finite element solution of engineering problems the main tasks of mesh generation, processing (calculations) and graphical representation of results are usually assigned to independent computer programs. These programs can either be embedded under a common shell (or interface) to enable the user to interact with all three parts in a single environment or they can be implemented as separate sections of a software package. Development and organization of graphics programs requires expertise in areas of computer science and software designs which outside the scope of text dealing with finite element techniques.

With finite-element program ANSYS, thermal conductivity analysis is carried out through the prepared composite body. A three-dimensional physical model with spheres-in-a-cube lattice array have been used to simulate the microstructure of composite materials for six different filler concentrations varying from 1vol% to 12 vol% and corresponding effective thermal conductivities of these composites are determined using ANSYS.

Steps involved in FEM:-

1. Defining the problem.

2. Selection of field variables and elements.

3. Modelling.

4. Discretize the elements (Meshing).

5. Apply boundary conditions.

6. Solve the system of equations to get nodal unknowns.

7. Post-processing of solution to get required values.

Description of the problem:-

The determination of effective properties of composite materials is of paramount importance for functional design and application of composite materials. Microstructure represents shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. The effect of microstructure on the effective properties can be gained from the investigation of composites with periodic structure.

For numerical analysis, the temperatures at the nodes along the surfaces ABCD is given as T_1 (=100^0C) and the convective heat transfer coefficient is given as 2.5 W/m^2K at ambient temperature of 27^0C. The heat flow is unidirectional with suitable boundary conditions are shown in Fig.5.3. The rest surfaces parallel to the direction of the heat flow are assumed heat flux to be zero. The temperatures at the nodes in the interior region and on the adiabatic boundaries are obtained with ANSYS programming.

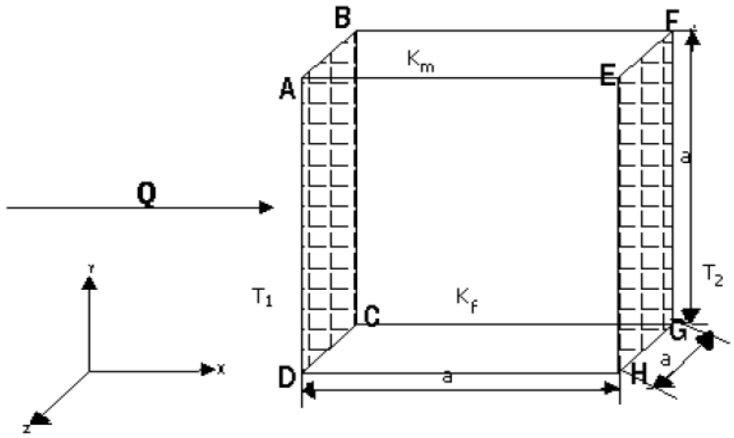

Fig.5.3: Boundary Conditions

Assumptions

The numerical analysis is based on the following assumptions:

1. The composites are **macroscopically homogeneous**.
2. Locally **both the matrix and filler** are homogeneous **and isotropic.**
3. The thermal contact resistance between the filler and the matrix is negligible.
4. The **composite lamina is free of voids.**
5. The problem is based on 3D physical model.
6. The filler are arranged in a square periodic array/uniformly distributed in matrix.

Table 5.2: Effective thermal conductivities obtained from different models and experimentation

Filler Content (Vol %)	Effective thermal conductivity of composites K_{eff}(W/m-K)							
	Rule of mixture	Geometric mean model	Maxwell's equation	Bruggeman's model	Lewis and Nielsen's equation	Proposed model	FEM simulation value	Experimental value
0	0.363	0.363	0.363	0.363	0.363	0.363	0.363	0.363
1.41	0.368	0.387	0.378	0.378	0.375	0.481	0.393	0.449
3.35	0.375	0.423	0.399	0.401	0.394	0.548	0.435	0.521
5.23	0.383	0.461	0.421	0.425	0.414	0.607	0.484	0.579
7.85	0.394	0.519	0.452	0.461	0.443	0.687	0.542	0.640
9.42	0.403	0.558	0.472	0.483	0.462	0.735	0.607	0.703
11.3	0.409	0.608	0.497	0.514	0.487	0.795	0.654	0.763

The effective thermal conductivity values of the particulate filled epoxy composites with varied proportions of alumina particulates obtained using Maxwell's correlation, ROM model, Lewis-Nielson Model and those obtained from FEM analysis are presented in Table5.2. It elaborates a brief comparison among the results obtained using these models with regard to the values of effective conductivity obtained experimentally.

Temperature Profiles of Composite from FEM Analysis:-

The temperature profiles obtained from FEM analysis for the composites with particulate concentrations of 1.4, 3.35, 5.23, 7.85, 9.04 and 11.3 vol. % are presented in Fig.5.4 (a-f) respectively.

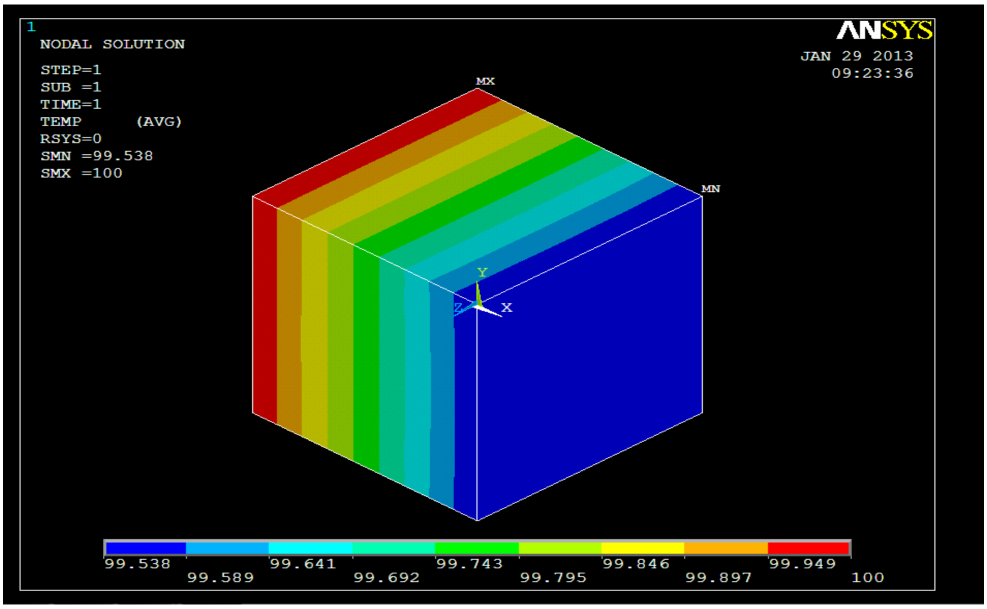

Fig 5.4(a): Temperature profiles for composite of filler concentration 1.42 vol %

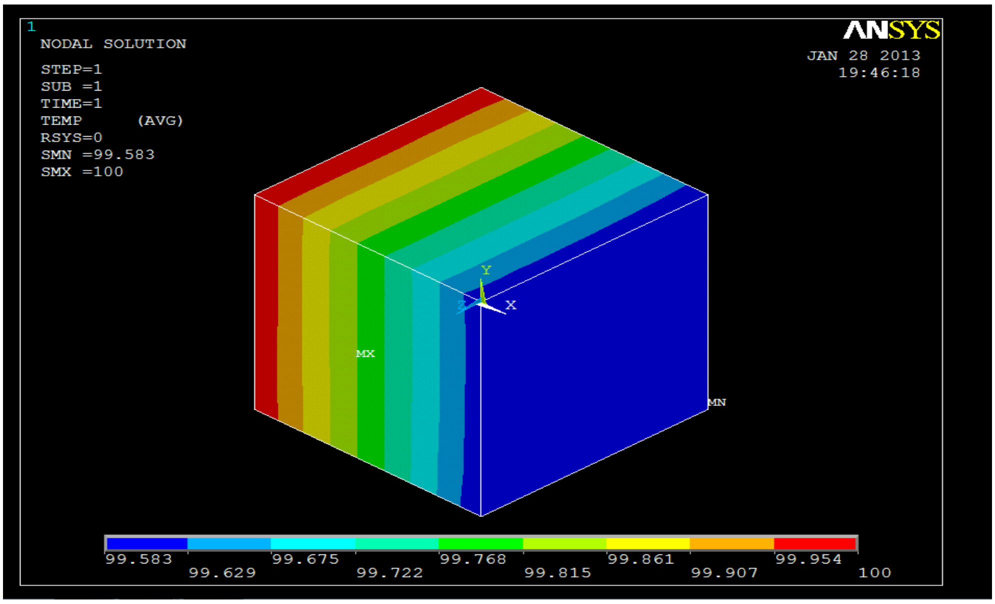

Fig 5.4(b): Temperature profiles for composite of filler concentration 3.35 vol %

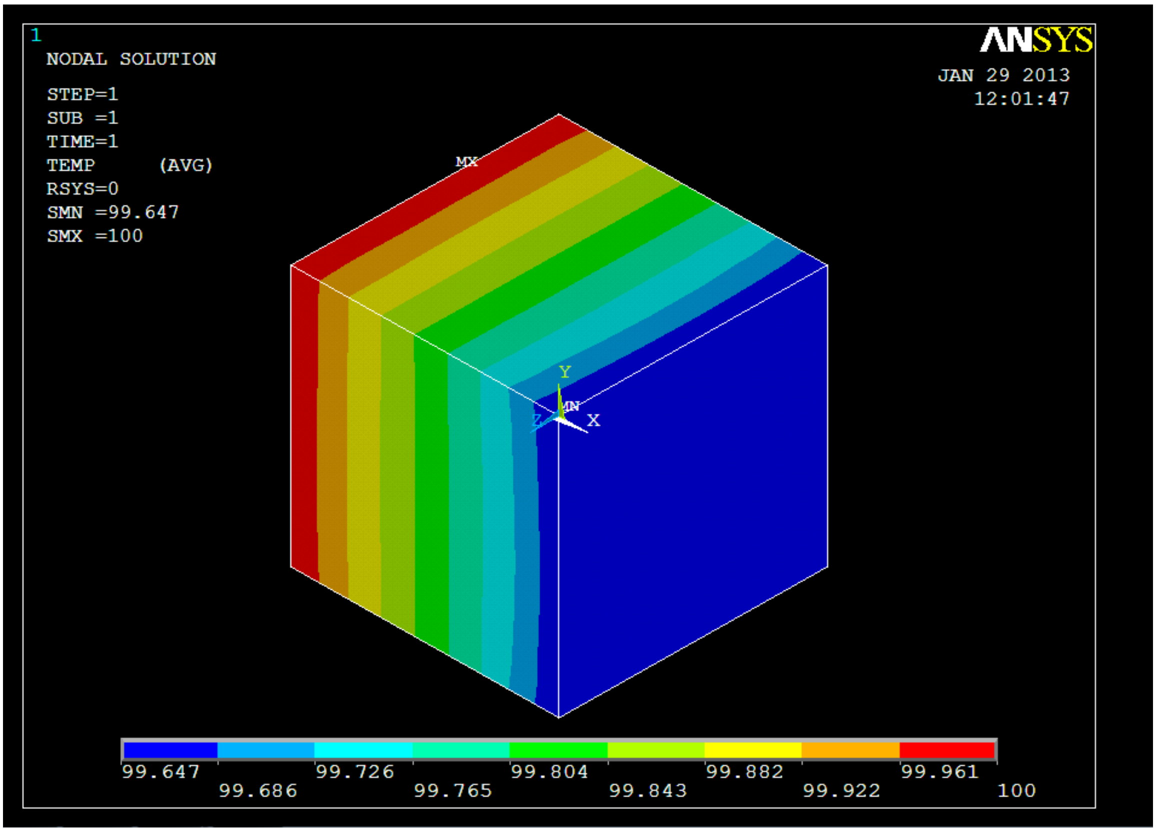

Fig 5.4(c): Temperature profiles for composite of filler concentration 5.23 vol %

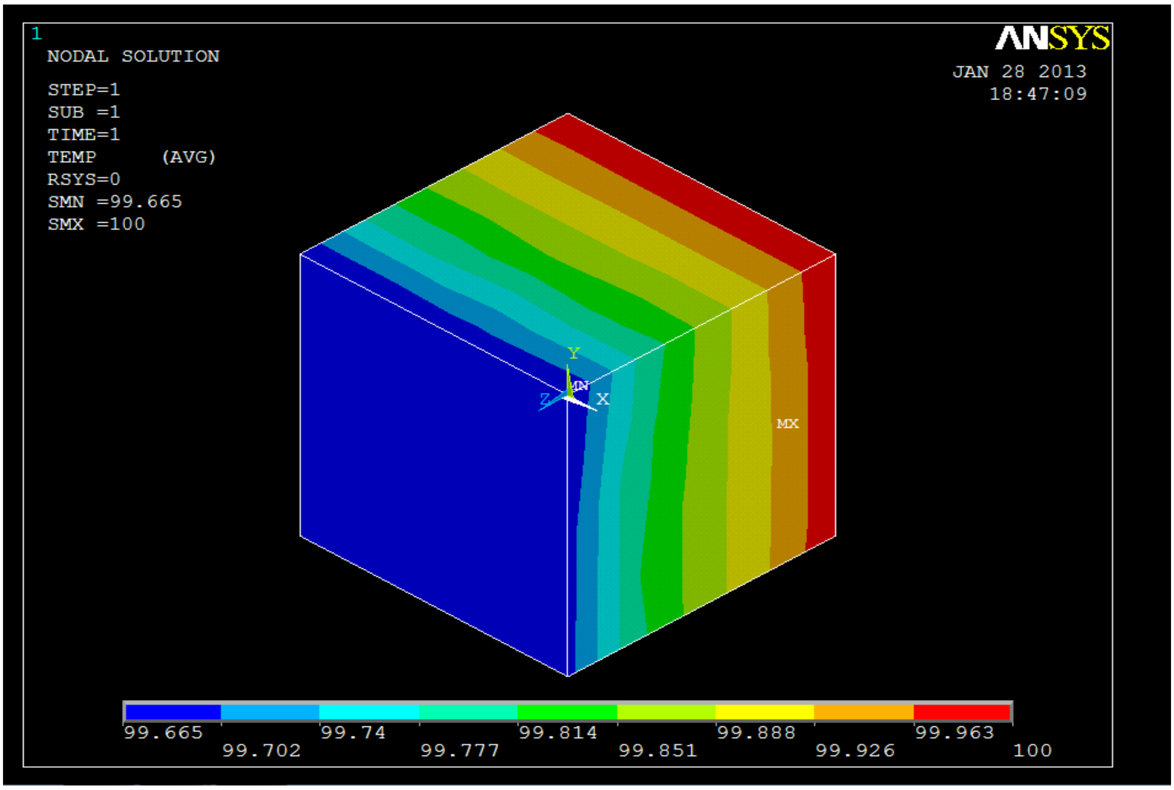

Fig 5.4(d): Temperature profiles for composite of filler concentration 7.85 vol %

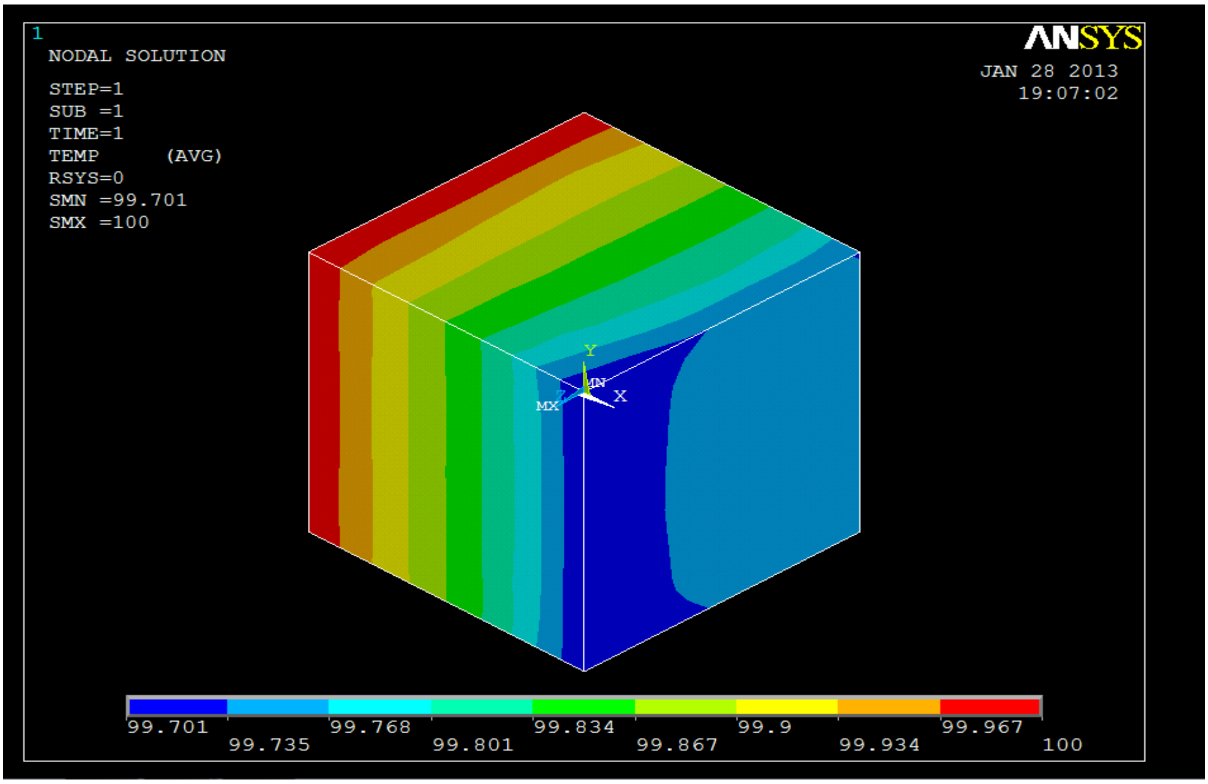

Fig 5.4(e): Temperature profiles for composite of filler concentration 9.4 vol %

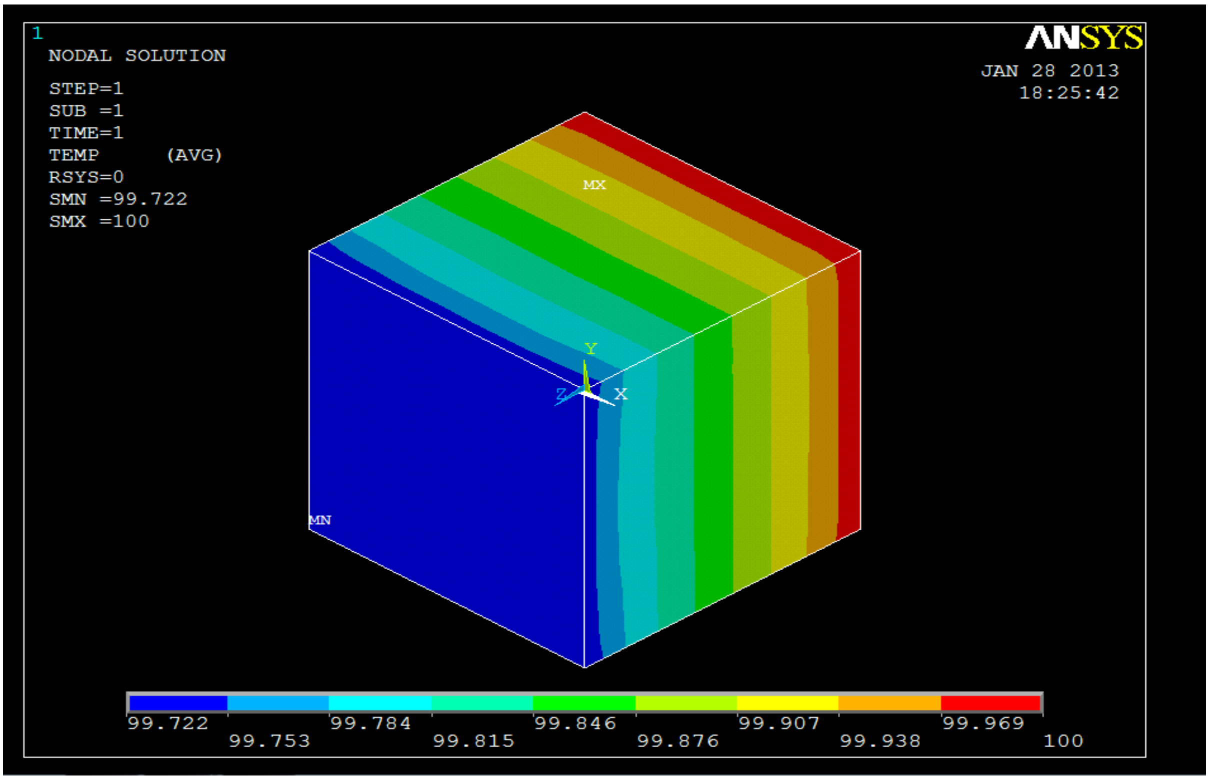

Fig 5.4(f): Temperature profiles for composite of filler concentration 11.3 vol %

The determination of effective properties of composite materials is of paramount importance for functional design and application of composite materials. One of the important factors that influence the effective properties and can be controlled to an appreciable extent is the microstructure of the composite. Here, microstructure means the shape, size distribution, spatial distribution and orientation distribution of the reinforcing inclusion in the matrix. Increase in filler concentrations attributes to the formation of continuous chains or network of conducting phase that spans through the insulating matrix. When the probability of the particles being in contact become large, highly conductive particles produce a path of low resistance for heat to flow and this would contribute to an increase in effective conductivity. Due to spherical nature of micro-sized alumina particles, the stress concentration effect is reduced which increases the contact between the particles resultingan increasein$K_{eff}$of system.

Table 5.3: Percentage error of different models with experimental values

Filler Content (Vol %)	% of errors with experimental values					
	Rule of Mixture	Geometric mean model	Maxwell's equation	Bruggeman's model	Lewis and Nielsen's equation	Proposed model
1.4	18.04	13.8	15.8	15.8	16.5	6.7
3.35	28.02	18.8	23.42	23.03	24.4	4.9
5.23	33.85	20.38	27.3	26.6	28.5	4.6
7.85	38.44	18.9	29.4	27.9	30.8	6.8
9.4	42.67	20.63	32.9	31.3	34.3	4.4
11.3	46.39	20.31	34.9	32.6	36.2	4.0

Table 5.4: Percentage error of FEM Analysis with experimental values

Sample	Volume (%)	Weight (%)	FEM (% errors)
1	1.4	4.78	12.4
2	3.35	10.91	16.5
3	5.23	16.345	16.4
4	7.85	23.15	15.3
5	9.4	26.84	13.7
6	11.3	31.05	14.3

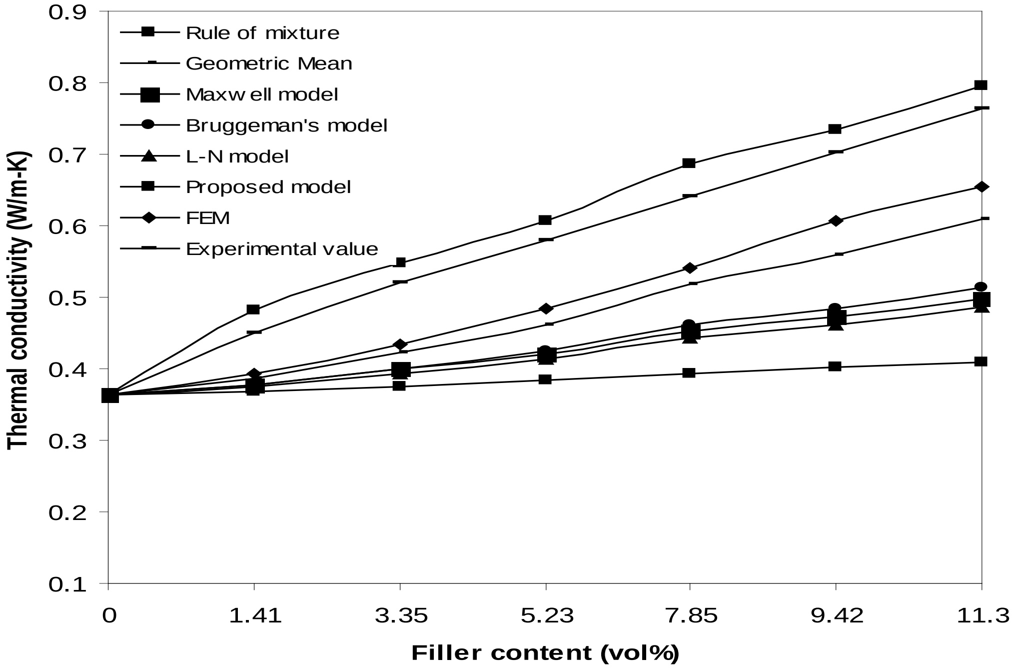

Fig 5.5: Comparison of values of K_{eff} for different theoretical models and FEM

It can be seen from the fig.5.5 that the K_{eff} increases non-linearly with increase in volume fraction of filler particles which agglomerates the neighbouring particles while for low filler concentrations there is low dispersion of particles in the matrix. Figure 5.5 clearly demonstrates that although the thermal conductivity of a composite increase with the increase of the filler content in the whole range of the filler content, the sensitivity of the thermal conductivity to the filler content is dependent on the filler content in the composite. The reason leading to this outcome is that for the composite with low filler loading, most filler powders are surrounded by the matrix and separated with each other which mean the channels for thermal conduction is mainly made up of the matrix but not fillers; in other words, the thermal conductivity of the composite is mainly dependent on the matrix. Therefore, in this situation, the thermal conductivity of the composite increases slowly with the increase of the filler content. Since heat tends to flow through the fillers just as electric current where electric resistance is low, the thermal conduction of fillers makes major contribution to the value of the thermal conductivity of the composites. Well-dispersed fillers are easy to become the component of the conductive channels; in addition, improved interface tends to increase the thermal conductivity by minimizing the phonon scattering at the interface. In case of the composite with higher filler content, the effect associating the surface treatment of filler enhances because there are more interfaces in the composite.

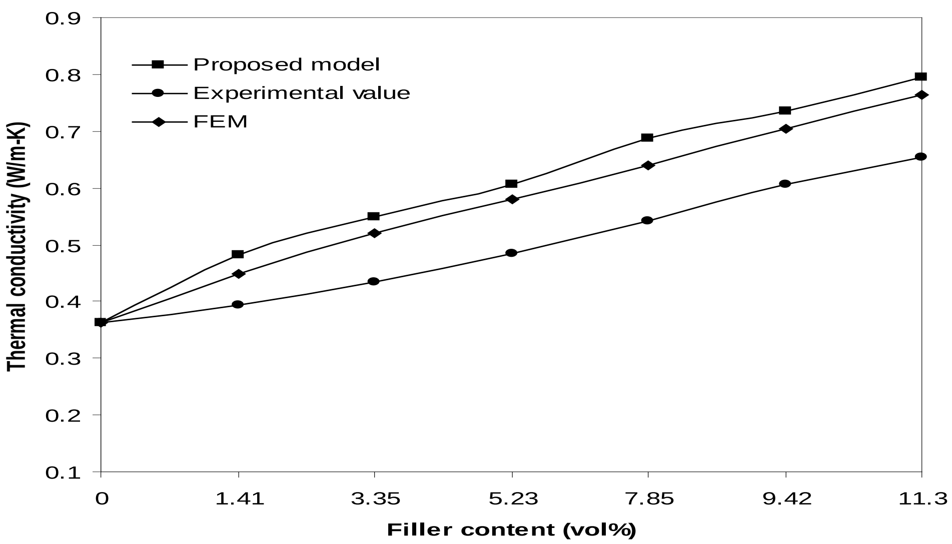

Fig.5.6: Comparison of K_{eff} using proposed, experimental and FEM values

The values obtained from theoretical models are quite low compared to the measured values, but variation of the theoretical estimation by proposed model of the effective thermal conductivity are well within the range of experimental measured values as shown in fig.5.6. A variation of about 7-8% is observed between the experimental and proposed model values which accounts to the assumptions taken while deriving the equation for the proposed model. The equation is derived on the basis of micro-spherical alumina particulates being uniformly distributed in the polymer matrix, but practically in a fabricated composite uniform distribution of particles is not possible as they are dispersed randomly. Also, it is assumed that heat flow is unidirectional which, in actual practice it is impossible to restrict the heat flow in one direction.

The simulated values of effective thermal conductivity of the composites obtained from FEA are presented in Table 5.2along with the corresponding measured values for different samples with filler concentration varying from 0vol% to 11.3vol%. Table 5.3 & 5.4 shows percentage errors established between thermal conductivity model values and proposed model values with the experimental values respectively. It is noticed that K_{eff}obtained from the proposed model are quite in agreement with experimental values than the FEM values, even though theerrors associated with sphere-in-cube simulations lies in the 0-17% which is quite reasonable. However, it leads to conclusion that for a particulate filled composite of this kind the FEM can be gainfully employed as a predictive tool in determining the effective thermal conductivity for a wide range of particle concentration. The difference between the simulated values and the measured value of conductivity may be attributed to the fact that some of the assumptions taken for the numerical analysis are not real, as in the fabricated composite sample, the fillers are actually dispersed in the resin almost randomly. With addition of 1.4 vol. % of Al_2O_3, the thermal conductivity increases by about 8.6 % and with addition of 11.3 vol.% of Al_2O_3, the thermal conductivity increases by about 40% when compared with neat epoxy resin. It might be that the aluminium oxide particles produce heat transfer links in the matrix due to the small distance between neighbouring particles in the case of high concentration and thus, the conductive capacity of the composite system is quickly enhanced.

CHAPTER 6

SUMMARY and CONCLUSION

Thesis Summary

As mentioned earlier, the continuing miniaturization of electronic devices and the increasing power output of electrical equipment have created new challenges in packaging and insulating materials. Dissipating adequate amount of heat from electronic devices is essential, otherwise, accumulated heat induces thermal fatigue that costs service life and operation efficiency. The key goals are to develop materials with high thermal conductivity, low coefficient of thermal expansion (CTE), low dielectric constant, high electrical resistivity, high breakdown strength, and most importantly, low cost. Polymeric materials have attracted increasing interest because of their excellent process ability and low cost; however, most polymers are thermally insulating and have a thermal conductivity between 0.1 and 0.5 W/m-K.Such thermal issues can be elevated using thermally conductive and electrically insulating ceramic fillers like aluminium nitride, aluminium oxide, silicon carbide and boron nitride doped in PMCs.

The research presented in this thesis consists of three parts:

- The first part describes the detailed fabrication of epoxy based composites filled with micro-sized aluminium oxide (Al_2O_3) particles in different volume proportions by hand lay-up technique.
- The second part provides information about physical characterization of the prepared composites which include comparison of theoretical and measured density. A SEM analysis is done to study the morphological structure of filled polymer composites.
- The third part includes an assessment of the effective thermal conductivity of these composites using finite element method followed by their validation with experimental calculated values and a brief comparison is done with developed theoretical model and existing thermal conductivity models proposed by the authors. Besides, thermal mechanical analysis is carried out that includes effect of filler content on Tg of composites and CTE simultaneously.

Conclusions

The numerical and experimental investigation on thermal conductivity of alumina particulates filled epoxy composites has led to specific conclusions:

- The addition of conductive fillers in the polymer matrix is an effective way to increase thermal conductivity of polymers for several industrial applications. To make adequate use of the filled polymer composites especially in printed circuit boards and micro-electronics applications, experimental results are necessary for the specific type of composite in question.
- Successful fabrication of epoxy composites filled Al_2O_3 particles by hand lay-up technique is possible.
- Finite element method (FEM) can be gainfully employed for determination of effective thermal conductivity of these composites with different amount of Al_2O_3content.
- Incorporation of micro-sized Al_2O_3 results in significant increase of thermal conductivity of pure epoxy resin and there by enhances its conduction capability. In comparison to neat epoxy resin, the thermal conductivity increases by about 40 % with addition of 11.3 vol% of Al_2O_3 from FEM analysis and around an increase of 80% with addition of 22.1 vol% of Al_2O_3 from experimental analysis.
- Higher percentage of Al_2O_3 will achieve more improved thermal conductivity and a rapid increase in thermal conductivity with increase in filler content is observed.
- Increase in Al_2O_3content in epoxy results in increase in the glass transition temperature and decrease in the value of CTE, hence providing a wider application range and superior thermal stability.
- With increased thermal conductivity, this new class of Al_2O_3filled epoxy composites can be used for applications such as electronic packages, encapsulations, printed circuit boards.

Scope for future work

This work leaves a wide scope for future investigators to explore many other aspects of thermal behaviour of particulate filled composites. Some recommendations for future research include:

- Effect of filler shape and size on thermal properties of the composites
- Exploration of new fillers and polymers for development of materials having high thermal conductivity and low electrical conductivity.

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